



August 8, 2005

Express Mail No. ED 999220622 US

MAIL STOP APPEAL BRIEF-PATENTS Commissioner for Patents P.O. Box 1450 Alexandria, VA 22313-1450

Re:

U.S. Patent Application Serial No. 10/618,394 for "Coupled Styrenic Block

Copolymers and Process for Preparing Same"

Inventor(s): Robert C. Bening, et al.

Filing Date: July 11, 2003 Attorney Docket No.: W-0006

Dear Sir:

Please find the following enclosed documents:

1. Appeal Brief (3 copies).

The Commissioner is hereby authorized to charge and credit KRATON Polymers U.S. LLC Deposit Account No. 50-1951 for the fees necessitated with the filing of this correspondence.

The confirmation from this facsimile will indicate receipt by the USPTO.

Respectfully submitted,

Muhal A. Masse

Michael A. Masse

Intellectual Property Asset Manager

PATENT

Atty. Dkt. No. W-0006

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

n re Application of:

Robert Bening et al.

erial No.:

10/618,394

Filed:

July 11, 2003

For:

Coupled Styrenic Block

Copolymers and Process for Preparing

Same

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Attorney Dkt. No.: W-0006

Group Art Unit: 1711

Examiner:

Asinovsky, Olga

Mail Stop Appeal Brief - Patents **Commissioner for Patents** P.O. Box 1450 Alexandria, VA 22313-1450

Dear Sir:

Certificate of Express Mail 37 CFR 1.10

I hereby certify that this paper or fee is being deposited with the U.S. Postal Service "Express Mail Post Office to Addressee" service under 37 CFR 1.10 on the date indicated below, addressed to: Mail Stop Appeal Brief - Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450. "Express Mail" Label No.: ED 999220622 US

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APPEAL BRIEF UNDER 37 CFR 1.192

Applicants submit this Appeal Brief to the Board of Patent Appeals and Interferences on appeal from the Final decision of the Examiner of Group Art Unit 1711 dated May 26, 2005. Please charge the amount for filing this brief (\$500) and any additional fees needed to make this submission timely to Deposit Account 50-1951(W-0006). Three copies of the brief are submitted for use by the Board.

Real Party in Interest

The present application is assigned to KRATON Polymers U.S. LLC.

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Related Appeals and Interferences

Applicants assert that no other appeal or interference is known to the Applicants, the Applicants' legal representative, or assignee which will directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

Status of Claims

Claims 11-18 and 22-34 remain pending in the application. Claims 11-18 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Schade et al. (U.S. Patent 6,506,846). Claims 22-34 stand rejected under 35 U.S.C. § 112, second paragraph. Claims 22-34 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Dillman et al. (U.S. Patent 5,420,203) in view of Schade et al. (U.S. Patent 6,506,846). The rejections of claims 11-18 and 22-34 are appealed. No claims have been allowed. The pending claims are shown in the attached Appendix.

Status of Amendments

The claims in the Appendix include amendments presented after first office action. Applicants filed a response to the final rejection in which claims 11, 22-24 and 26 were amended. Applicants' amendment after final rejection has not been entered.

Summary of the Invention

One embodiment of the present invention is a method to make radial styrenic block copolymers. The inventive method comprises anionic polymerization of styrenic and dienic monomers to produce a living polymer, addition of a metal alkyl compound during or after polymerization, and addition of a diester coupling agent to couple and thereby produce the radial polymer (paragraph [0010] of the application as filed and Claim 11). One essential feature of the method is the addition of the metal alkyl during or after polymerization. This novel method yields the polymer composition which will be subsequently discussed.

A second embodiment of the present invention is the radial styrenic block copolymer produced by the above method. The inventive block copolymer has the general formula (AB)nX where A is a styrenic block, B is a dienic block, X is the residue of a diester coupling agent, n is the number of arms bonded to the coupling agent, and

the weight percentage of the polymer fraction having n equal to at least 5 is less than about 8 percent (paragraphs [0009] and [0028] of the application as filed and Claim 22). One essential feature of the block copolymer is the low amount of radial polymer having 5 or more arms. This polymer is only obtainable by the novel method of Claim 11.

Issues Presented

- I. Whether claims 11 18 are unpatentable under 35 U.S.C. § 103(a) over U.S. Patent 6,506,846.
- II. Whether claims 22 34 are unpatentable under 35 U.S.C. § 112, second paragraph, as being indefinite, for failing to particularly point out and distinctly claim the subject matter which the Applicants regard as the invention.
- III. Whether claims 22 34 are unpatentable under 35 U.S.C. § 103(a) over U.S. Patent 5,420,203 in view of U.S. Patent 6,506,846.

Grouping of Claims

Claims 11 - 18 (Group I) stand or fall together for the arguments presented by the Applicants where Claim 11 is representative of the Applicants' arguments.

Claims 22 - 34 (Group II) stand or fall together for the arguments presented by the Applicants where Claim 22 is representative of the Applicants' arguments.

Arguments

1. 35 U.S.C. § 103(a) rejection of Claims 11 – 18.

The Examiner erred in rejecting claims 11 – 18 under 35 U.S.C. § 103(a) as being unpatentable over U.S. Patent 6,506,846 (Schade et al.) since Schade et al. fail to teach or suggest the addition of metal alkyls during or after polymerization but before coupling and necessarily before termination. In other words, Schade et al. fail to tech or suggest that the metal alkyl <u>must</u> be present during the reaction of the living polymer and the coupling agent and certainly before termination. Therefore, the Examiner has not established a *prima facie* case of obviousness.

Schade et al. teach a process for preparing impact modified thermoplastics comprising anionically polymerizing a rubber followed by coupling or termination which in turn is followed by initiation of an anionically polymerizable monomer by a metal alkyl compound. The object of Schade et al. is to provide a method to make impact-modified thermoplastic molding compositions which comprise a soft phase made from a rubber dispersed in a hard matrix composed of vinyl aromatic monomers (col. 2, lines 9 - 16). In the two-step method of Schade et al. the rubber is polymerized anionically in the presence of styrene and then the hard matrix is formed in a second step (col. 3, lines 29 – 33). The first step comprises polymerization of the rubber using alkali metals, coupling and termination to complete the formation of the rubber polymer (col. 3, line 61 - col. 4, line 8 and examples 1 - 8). The second step comprises initiation and polymerization of the hard matrix. Schade et al. teach that metal alkyls may be used as "initiators" in this second step to form the thermoplastic matrix polymer (column 4, lines 28 – 37 and examples 9 – 11). A mixture of a rubber polymer and a thermoplastic polymer are made by the method of Schade et al. It is for initiation of the thermoplastic matrix polymer, which is never coupled, that the metal alkyl is used.

Importantly, Schade et al. teach that while the metal alkyls can initiate polymerization they "otherwise have only a retarding effect" (column 4, lines 33 – 35). Thus, Schade et al. teach that the metal alkyl has only two effects: initiation and retardation. What is meant by initiation and retardation is taught by Schade et al. The specification (col. 4, lines 28 - 43) and the examples (examples 9 - 11) demonstrate that initiation of polymerization of a hard matrix by metal alkyls happens in a separate, second step after polymerization and termination of a rubber comprising styrenic and diene monomer units. Initiation serves to polymerize the hard matrix which is to embed the rubber. Retardation is the means to solve the problem of very fast polymerization leading to high heat generation (col. 1, lines 46 - 50). Schade et al. only teach the initiation of polymerization by addition of metal alkyls or the slowing down of a

polymerization to reduce heat generation. Schade et al. specifically state that these are the only two effects of the metal alkyl (column 4, lines 33 - 35).

Claim 11 of Appellants' invention recites a method for preparing a radial styrenic block copolymer comprising (a) contacting styrenic and dienic monomers with an anionic polymerization initiator which is an organo-substituted alkali metal compound in a suitable solvent to form a living polymer cement, (b) adding from about 0.01 to about 1.5 equivalents of a metal alkyl compound per equivalent of living polymer chain ends to the cement, during or after polymerization, wherein the alkyl groups of the metal alkyl compound are chosen so that they will not exchange with the living polymer chain ends and the metal alkyl compound is selected from the group consisting of aluminum, zinc and magnesium alkyls having from 1 to 20 carbon atoms per alkyl substituent, and (c) adding a diester coupling agent to the cement under reaction conditions sufficient to couple the living polymer.

The object of this embodiment of Appellants' invention is to provide a method for producing block copolymers having increased three and four arm contents and reduced five and higher arm contents (paragraph [0008] of the application as filed). It is well known in the art that the coupling process is not limited to making block copolymers having three or four arms but can include coupled polymers having a larger number of arms. The Appellants' method, which reduces the fraction of coupled polymers having five or more arms, yields block copolymers having reduced viscosities. This is beneficial for block copolymer processing in general and applications such as bitumen modification in particular (paragraph [0007] and the examples of the application as filed).

One essential difference between Appellants' method and the method of Schade et al. is in the time of addition of the metal alkyl. Appellants' method requires addition of the metal alkyl during or after polymerization but before coupling and necessarily before termination. This leads to the effect of reducing the fraction of coupled polymer having five or more arms. The method of Schade et al. requires addition of the metal alkyl to start the second polymerization after termination of the anionic polymerization of the rubber

phase. In Schade et al. the metal alkyl is <u>never</u> used in the anionic polymerization or coupling of the rubber.

Schade et al. teach that besides initiation, metal alkyls have only a retardation effect. (col. 4, lines 33 – 35). Therefore, Schade et al. do not teach and cannot suggest a modification to reach the method discovered by Appellants. Appellants' method involves coupling in the presence of metal alkyl and this affects a narrow molecular weight distribution of the coupled polymer (see Table 1 of Appellants' application as filed). Appellants' coupling process is entirely different and separate from initiation and retardation. There is no motivation in Schade et al. to modify the method to the Appellants' method. Further, there is no suggestion in the prior art that the method of Schade et al. should be modified to achieve the Appellants' method. A *prima facie* case of obviousness cannot be established absent some teaching, suggestion or incentive supporting the modification (*In re Gordon* 773, F.2d 900, 902, 221 U.S.P.Q 1125, 1127 (Fed. Cir. 1984); *In re Mills* 916 F.2d at 682, 16 U.S.P.Q.2d at 1432 (Fed. Cir. 1990); MPEP 2143.01).

Furthermore, the modification required to achieve the Appellants' method would render the method of Schade et al. inoperable. Appellants rely upon the teaching of Schade et al. (column 4, lines 28 – 35). Therein, it is clearly stated that the method of Schade et al. only works if use is made of a rubber solution which has been 1) prepared by anionic polymerization, and 2) terminated by chain termination or coupling. Only in this case do the metal alkyl compounds initiate the polymerization of the hard matrix. In the Appellants' method the metal alkyl must be added during or after polymerization but before the coupling step. The metal alkyl must be present during the reaction of the living polymer and the coupling agent and certainly before termination. In the method of Schade et al. the metal alkyl must be added after termination of the rubber when the polymer is no longer living. According to this teaching, addition of the metal alkyl before termination and still in the presence of living anionic chains and alkali metal would not serve to initiate the second step of Schade et al. For this further reason, Schade et al. cannot provide the suggestion or motivation to

modify the method since the modification would render the invention of Schade et al. unsatisfactory for its intended purpose and as such teaches away from Appellants' inventive method (*In re Gordon* 773, F.2d 900, 221 U.S.P.Q 1125, (Fed. Cir. 1984); MPEP 2143.01).

II. 35 U.S.C. § 112, second paragraph, rejection of Claims 22 – 34.

The Examiner erred in rejecting claims 22 – 34 under 35 U.S.C. § 112, second paragraph, as being indefinite for failing to point out and distinctly claim the subject matter which Applicants regard as the invention since there is clearly sufficient antecedent basis for the limitation in claim 22.

Claim 22 recites a radial block copolymer produced by the method of claim 11. Claim 11 recites a method for producing a radial styrenic block copolymer. The Examiner has noted in the Office Action dated February 7, 2005 that claim 22 recites the limitation "wherein said copolymer is produced by the method of claim 11" and asserts there is insufficient antecedent basis for this limitation in claim 22. The Examiner concedes, in the paper of February 7, 2005, that claim 11 discloses a method of making a radial styrenic block copolymer. Further, the Examiner concedes that the method of claim 11 includes a step(b) of adding a metal alkyl compound. However, the Examiner rejects claim 22 and in doing so states that claim 22 does not require a metal alkyl and the polymer of claim 22 is different from one produced by the method of claim 11. The Examiner errs in this assertion. Claim 22 recites a polymer made by, and including all the limitations of, the method of claim 11.

Appellants note that a claim which makes reference to a preceding claim to define a limitation is an acceptable claim construction which should not necessarily be rejected as improper or confusing under 35 U.S.C. 112, second paragraph (MPEP 2173.05 (f)). Further, a claim to a composition of matter may contain a reference to the process in which it is intended to be used without being objectionable under 35 U.S.C. § 112, second paragraph, so long as it is clear that the claim is directed to the product and not the process (MPEP 2173.05 (p)). Appellants' claim 22 lays out the elements of the claimed radial

styrenic block copolymer "wherein said copolymer is produced by the method of claim 11". Further, claim 11 lists all the essential steps in the method required to make the polymer of claim 22. Appellants assert that there is nothing confusing about this reference. The block copolymer of claim 22 is made by the method of claim 11.

Appellants maintain that claim 22 has been improperly rejected by the Examiner under 35 U.S.C. § 112, second paragraph. Claims 23 - 24 are rejected because of their dependency on claim 22: Appellants therefore assert that claims 23 - 24 have also been improperly rejected by the Examiner.

III. 35 U.S.C. § 103(a) rejection of Claims 22 – 34.

The Examiner erred in rejecting claims 22 – 34 under 35 U.S.C. § 103(a) as being unpatentable over U.S. Patent 5,420,203 (Dillman et al.) in view of U.S. Patent 6,506,846 (Schade et al.) since Dillman et al. fail to teach or suggest metal alkyls or their use for any purpose and Schade et al., while using metal alkyls, fail to provide the motivation to modify the method of Dillman et al. and produce a polymer having the distribution of couples species produced by Appellants. The Examiner has not established a *prima facie* case of obviousness.

Dillman et al. teach a method comprising (a) anionically polymerizing at least one vinyl aromatic hydrocarbon and at least one conjugated diene to form a living block copolymer, (b) coupling the block copolymer with a coupling agent selected from the group consisting of esters of carboxylic acids and asymmetric silanes to create a coupled polymer, and (c) heating the composition to a temperature sufficient to decouple the coupled copolymer such that the amount of uncoupled copolymer in the composition ranges from 10 to 100% by weight of the total amount of block copolymer. The object of Dillman et al. is to provide a process for producing a composition which contains block copolymers having low cohesive strength and low elasticity (col. 1, lines 8-10). The radial block copolymer is represented by the formula (A-B)nX (col. 2, lines 45-47). Dillman et al., however, are silent with regard to arm number distribution of the

coupled polymer. Further, Dillman et al. are silent with regard to metal alkyls or their use for any purpose.

Schade et al. have been discussed previously. Schade et al. do not teach a method wherein metal alkyls are used in a coupling process. Schade et al. do teach that metal alkyls only have a retardation effect and an initiation effect and, therefore, teach away from any effect of metal alkyls on arm number distribution during coupling.

Claims 22 - 34 of Appellants' invention claim a coupled, radial polymer having less than about 8% wt of the polymer is such that "n is at least 5". The coupling reaction of the present invention, like all other coupling reactions, does not yield a single species. There will be uncoupled arms (n = 1), two arms coupled together (n = 2), three arms coupled together (n = 3) and so on, all making up the coupled polymer composition. It is clear from the Applicants' discussion of the coupling process (paragraphs [0021] - [0023] and in Table 1 of the Examples in application as filed) that an arm number distribution results from the coupling reaction.

Table 1 of Appellants' application shows the distribution of coupled species resulting from Appellants' method of coupling in the presence of metal alkyls. Examples 1-5 show that when metal alkyls are present in a metal alkyl lithium ratio of about 1 the amount of "n is at least 5" (expressed as "n = 5+") is equal to or less than 5.3 wt%. Comparative Example 6 shows that when metal alkyls are absent during coupling an arm number distribution results in which the n = 5+ makes up 12.1 wt%. This is 2 to 5 times higher, a significant increase over the n = 5+ amount achieved in the present invention. When metal alkyls are present during coupling, inventive arm number distributions with n = 5+ less than about 8 wt% are achieved. The benefit of the block copolymer having n = 5+ less than about 8 wt% is generally presented in paragraph [0007] of the present application where it is said that the compositions having only n = 5 have a substantially greater viscosity than those having only n = 3. Increases in viscosity are not desirable.

The coupled block copolymer of Dillman et al. corresponds to Appellants Comparative Example 6 since Dillman et al do not employ a metal alkyl in the coupling process. Appellants' invention is different from Dillman et al. in the arm number distribution. Appellants' block copolymer has a reduced fraction of higher arm number species and therefore has the inherent benefit of lowered viscosity and easier processing.

There is no suggestion to modify the method of Dillman et al. to change the arm number distribution of the coupled polymer in any way or by any fashion or to produce a coupled polymer other than that produced by an unmodified coupling process. Nor is there any suggestion in Dillman et al. that arm number distribution, other than the ones obtained therein, are desirable or advantageous. Therefore, motivation to modify the coupling method to produce the Appellants' coupled polymer is absent. Because Schade et al. teach that metal alkyls only have a retardation effect and an initiation effect it teaches away from any effect of metal alkyls on arm number distribution during coupling. In fact, the teaching of Schade et al., where the metal alkyl is not present during coupling, would yield the same result of Comparative Example 6 with n = 5+ about or greater than 12 wt%. Therefore, Schade et al. provid no motivation to apply metal alkyls to the coupling process of Dillman et al. to make the Appellants' coupled polymer. A prima facie case of obviousness cannot be established absent some teaching, suggestion or incentive supporting the modification (In re Gordon 773, F.2d 900, 902, 221 U.S.P.Q 1125, 1127 (Fed. Cir. 1984); In re Mills 916 F.2d at 682, 16 U.S.P.Q.2d at 1432 (Fed. Cir. 1990); MPEP 2143.01).

Neither Dillman et al. nor Schade et al., considered alone or in combination, suggest or motivate one of ordinary skill in the art at the time of the invention to make a coupled polymer having a reduced fraction of coupled polymers where "n is at least 5" is less than 8% wt. The Examiner has erred in asserting a *prima facie* case of obviousness.

IV. Reasons why Appellants regard the Claims of the two groups to be separately patentable.

Appellants regard the claims of the two groups to be separately patentable because they are drawn to different inventions: Group I claims read on a method for making a block copolymer, represented by independent claim 11; Group II claims read on a block copolymer made by the process of Group I claims, represented by independent claim 22. Further, both groups have been separately regarded by the Examiner in the rejections. Group I is rejected under section 103(a) as being unpatentable over U.S. Patent 6,506,846. Group II is rejected under section 112, second paragraph, and under section 103(a) as being unpatentable over U.S. Patent 5,420,203 in view of U.S. Patent 6,606,846.

Conclusion

The Examiner has not established a *prima facie* case of obviousness against Appellants' claims 11 – 18. Appellants respectfully request reversal of the 35 U.S.C. § 103(a) rejection and allowance of claims 11 – 18 presented herein.

The Examiner has improperly rejected Appellants' claims 22 – 34 under 35 U.S.C. § 112, second paragraph and has not established a *prima facie* case of obviousness against these claims under 35 U.S.C. § 103(a). Appellants respectfully request reversal of these rejections and allowance of claims 22 – 34 presented herein.

Respectfully submitted,

August 8, 2005

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APPENDIX

The Pending Claims:

11. A method for preparing a radial styrenic block copolymer of Claim 1 comprising:

- (a) contacting styrenic and dienic monomers with an anionic polymerization initiator which is an organo-substituted alkali metal compound in a suitable solvent to form a living polymer cement;
- (b) adding from about 0.01 to about 1.5 equivalents of a metal alkyl compound per equivalent of living polymer chain ends to the cement, during or after polymerization, wherein the alkyl groups of the metal alkyl compound are chosen so that they will not exchange with the living polymer chain ends and the metal alkyl compound is selected from the group consisting of aluminum, zinc and magnesium alkyls having from 1 to 20 carbon atoms per alkyl substituent; and
- (c) adding a diester coupling agent to the cement under reaction conditions sufficient to couple the living polymer.
- 12. The method of Claim 11 wherein 0.9:1 to 1.1:1 equivalents of a metal alkyl compound per equivalent of living polymer chain ends is added to the cement.
- 13. The method of Claim 11 wherein the metal alkyl compound is a trialkyl aluminum compound.
- 14. The method of Claim 13 wherein the trialkyl aluminum compound is triethyl aluminum.
- 15. The method of Claim 1 wherein the molar ratio of diester to living polymer chains is from about 0.2:1 to about 0.3:1.
- 16. The method of Claim 11 wherein the molar ratio of diester to living polymer chains is about 0.25:1.
- 17. The method of Claim 11 wherein the metal alkyl compound is added in step (b) at or after 70 weight percent conversion of the monomers.
- 18. The method of Claim 17 wherein the metal alkyl compound is added in step (b) at or after 90 weight conversion of the monomers.

22. A radial styrenic block copolymer having a general formula:

 $(AB)_nX$

wherein:

- (i) A is a styrenic block,
- (ii) B is a dienic block,
- (iii) X is a residue of a diester coupling agent,
- (iv) n is the number styrenic block copolymer arms bonded to the residue of a diester coupling agent,
- (v) the molecular weight of the styrenic block copolymer arm (AB) is from about 2000 daltons to about 300,000 daltons, and
- (vi) the weight percentage of the polymer wherein n is at least 5 is less than about 8 percent, and

wherein said copolymer is produced by the method of Claim 11.

- 23. The radial styrenic block copolymer of Claim 22 wherein the weight percentage of the polymer wherein n is at least 5 is less than about 6 percent.
- 24. The radial styrenic block copolymer of Claim 23 wherein the weight percentage of the polymer wherein n is at least 5 is less than about 5 percent.
- 25. The radial styrenic block copolymer of Claim 22 wherein the weight percentage of the polymer wherein n=2 is less than about 5 percent.
- 26. The radial styrenic block copolymer of Claim 25 wherein the weight percentage of the polymer wherein n=2 and n is at least 5 is less than about 12 percent.
- 27. The radial styrenic block copolymer of Claim 22 wherein the styrenic block (A) is polystyrene.
- 28. The radial styrenic block copolymer of Claim 22 wherein the dienic block (B) is selected from the group consisting of polybutadiene, polyisoprene and mixtures thereof.
- 29. The radial styrenic block copolymer of Claim 22 wherein the molecular weight of the styrenic block copolymer arm (AB) is from about 3,000 daltons to about 150,000 daltons.

30. The radial styrenic block copolymer of Claim 26 wherein the molecular weight of the styrenic block copolymer arm (AB) is from about 30,000 daltons to about 100,000 daltons.

- 31. The radial styrenic block copolymer of Claim 22 wherein the residue of a diester coupling agent is a residue of a diester selected from the group consisting of dimethyl adipate, diethyl adipate, dimethyl terephthalate, diethyl terephthalate, and mixtures thereof.
- 32. A modified bitumen comprising an admixture of a radial styrenic block copolymer of Claim 22 and bitumen.
- 33. A hydrogenated radial styrenic block copolymer prepared by hydrogenating a radial styrenic block copolymer of Claim 22.
- 34. The hydrogenated radial styrenic block copolymer of Claim 33, wherein the radial styrenic block copolymer of Claim 22 is hydrogenated using a selective hydrogenation process.